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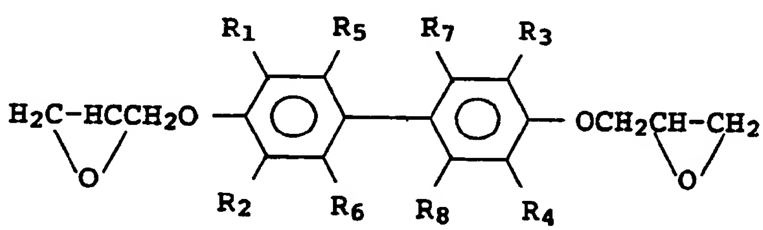
(54) Epoxy resin composition based on diglycidylether of biphenyldiol.

(57) Disclosed is an epoxy resin composition for sealing semiconductors which comprises as essential components:

(A) an epoxy resin containing a biphenyl type epoxy resin represented by the following formula (1) in an amount of 30-100% by weight on the basis of the total amount of the epoxy resin,



(1)

wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups,

(B) a hardener containing a phenolic resin hardener represented by the following formula (2a) or a naphthol resin hardener represented by the following formula (2b) in an amount of 30-100% by weight on the basis of the total amount of the hardener,

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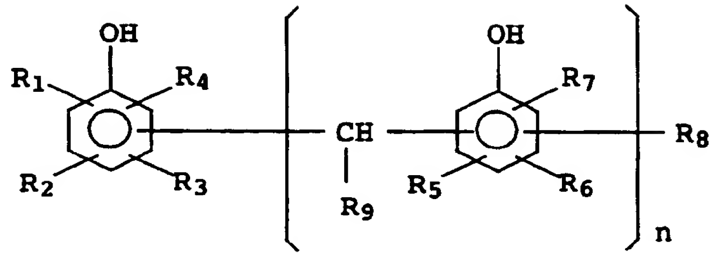
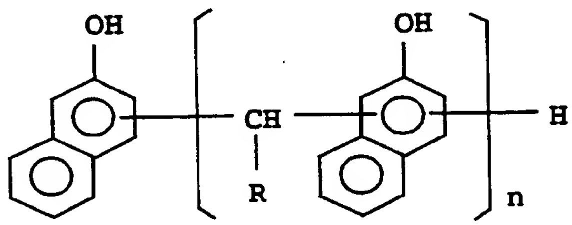


(2a)

wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups, R_9 is a group selected from phenyl, naphthyl, anthracenyl and alkyl groups, and n is 1-6,



(2b)

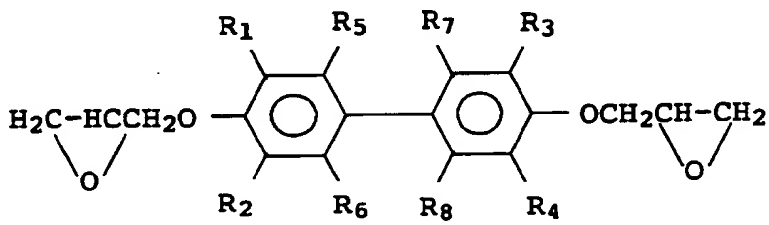
wherein R is a group selected from phenyl, naphthyl, anthracenyl and alkyl groups, and n is 1-6,
(C) an inorganic filler, and
(D) a hardening accelerator.

The present invention relates to epoxy resin compositions for sealing semiconductors which are excellent in soldering stress resistance in surface mounting of semiconductor devices.

Hitherto, electronic parts such as diodes, transistors and integrated circuits have been sealed with thermosetting resins. Especially, for sealing of integrated circuits, there have been used epoxy resin compositions prepared by hardening o-cresol novolak epoxy resins excellent in heat resistance and moisture resistance with novolak type phenolic resins. However, with recent increase in integration density of integrated circuits, use of large chips has gradually increased and the packages have been changed from the conventional DIP type to surface mounted small and thin QFP, SOP, SOJ and PLCC.

That is, large chips are sealed in a small and thin package and in this case, the stress causes formation of cracks and these cracks result in serious problems such as reduction of moisture resistance. Especially, the package is abruptly exposed to high temperatures of higher than 200 °C at the soldering step and this brings about the above problems. Thus, development of resin compositions which are suitable for sealing large chips and are high in reliability, has been desired.

As epoxy resins for solving the above problems, use of epoxy resins represented by the following formula (1) was proposed [cf. Japanese Patent Kokai (Laid-Open) No. 64-65116].



(1)

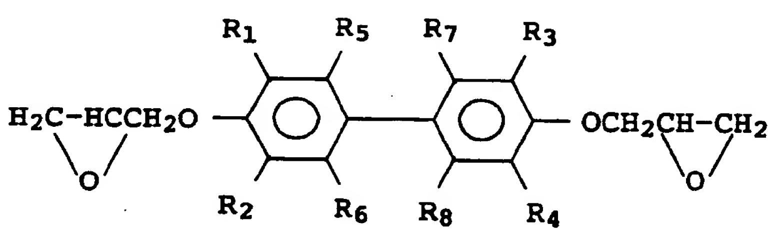
wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups.

By using the epoxy resins represented by the formula (1), it was attained to reduce the viscosity of the resin composition, and accordingly, it was attained to improve the soldering stress resistance by reducing the thermal expansion and the water absorption of the composition after molded by further increasing the amount of fused silica powders to be added to the composition. However, the increase of modulus resulting from increase in the amount of fused silica powders is also one problem, and further improvement of the soldering stress resistance is needed.

The object of the present invention is to overcome such problems, that is, to provide an epoxy resin composition used for sealing of semiconductors which is markedly improved in the soldering stress resistance when used for semiconductor packages in mounting on substrates by utilizing synergistic effects of reduction in water absorption and in linear expansion attained using the epoxy resin represented by the formula (1) and reduction in water absorption and impartation of flexibility attained using a specific hardener.

The epoxy resin composition of the present invention is an epoxy resin composition for sealing semiconductors which contains as essential components:

(A) an epoxy resin containing a biphenyl type epoxy resin represented by the following formula (1) in an amount of 30-100% by weight on the basis of the total amount of the epoxy resin,

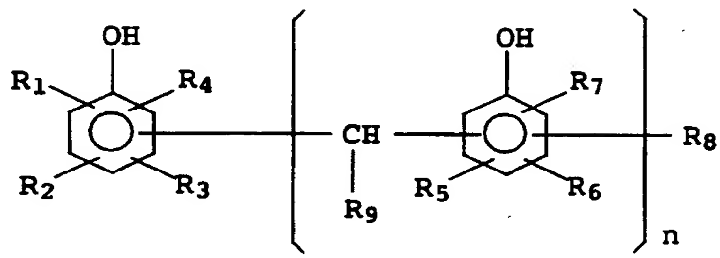


(1)

wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups,

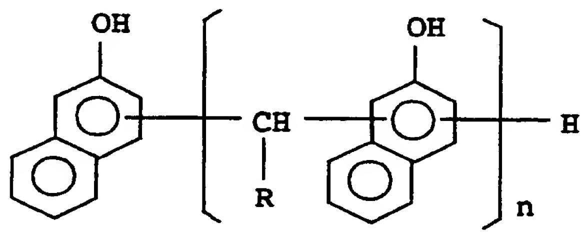
(B) a hardener containing a phenolic resin hardener represented by the following formula (2a) or a naphthol resin hardener represented by the following formula (2b) in an amount of 30-100% by weight on

the basis of the total amount of the hardener,



(2a)

wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups, R_9 is a group selected from phenyl, naphthyl, anthracenyl and alkyl groups and n is 1-6,



(2b)

wherein R is a group selected from phenyl, naphthyl, anthracenyl and alkyl groups and n is 1-6,
(C) an inorganic filler, and
(D) a hardening accelerator.

The epoxy resin composition of the present invention is markedly superior to the conventional epoxy resin compositions in the soldering heat resistance.

The epoxy resin represented by the structure of the formula (1) is a bifunctional epoxy resin having two epoxy groups in one molecule and is lower in melt viscosity and superior in fluidity at transfer molding as compared with the conventional polyfunctional epoxy resins. Therefore, fused silica powders can be added in a large amount to the composition and decrease in thermal expansion and water absorption can be attained and thus, epoxy resin compositions excellent in soldering stress resistance can be obtained.

The soldering stress resistance can be improved by adjusting the amount of the epoxy resin used. In order to obtain the effect to improve the soldering stress resistance, it is desired to use the biphenyl type epoxy resin represented by the formula (1) in an amount of 30% by weight or more, preferably 60% by weight or more on the basis of the total amount of the epoxy resin. If the amount is less than 30% by weight, decrease in thermal expansion and water absorption cannot be attained and the soldering stress resistance is insufficient. Furthermore, R_1-R_4 in the formula are preferably methyl groups and R_5-R_8 are preferably hydrogen atoms.

When other epoxy resins are used in combination with the epoxy resin represented by the formula (1), the other epoxy resins include the general polymers having epoxy group. Examples of them are bisphenol type epoxy resins, cresol novolak type epoxy reins, phenol novolak type epoxy resins, trifunctional epoxy resins such as triphenol methane type epoxy resins and alkyl-modified triphenol methane type epoxy resins, and triazine ring-containing epoxy resins.

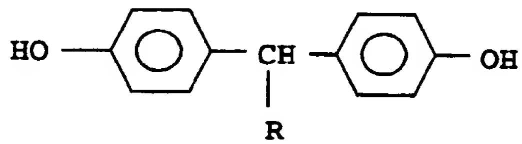
By using the hardener represented by the molecular structure of the formula (2a) or (2b), reduction in modulus and improvement of adhesion between lead frames and semiconductor chips at the soldering temperature can be attained and besides, decrease in thermal expansion and water absorption can be attained as compared with the conventional phenol novolak resin hardeners. Therefore, the hardeners used in the present invention are effective for reduction of the stress generated by thermal shock at the time of soldering and for inhibition of peeling of semiconductor chips caused by the stress.

The soldering stress resistance can be improved by adjusting the amount of the hardener represented by the formula (2a) or (2b). In order to bring about the effect of the soldering stress resistance, it is desired to use the hardener represented by the formula (2a) or (2b) in an amount of 30% or more, more preferably 60% by weight or more on the basis of the total amount of the hardener. If the amount is less than 30% by

weight, decrease in water absorption and modulus and adhesion between lead frames and semiconductor chips are insufficient and improvement in soldering stress resistance cannot be expected. Furthermore, the value of n is necessary to be in the range of 1-6. If n exceeds 6, fluidity at transfer molding decreases and moldability tends to deteriorate.

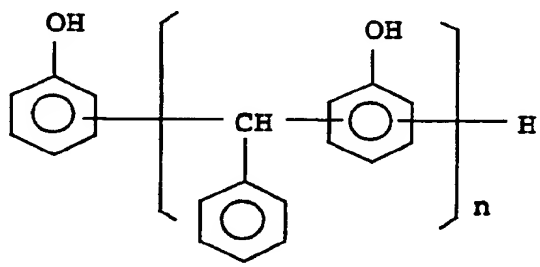
Furthermore, R_1-R_8 in the formula (2a) are preferably hydrogen or methyl groups and R_9 is preferably naphthyl group or an alkyl group.

Especially preferably, the formula (2a) is represented by the following formula (2a-1), (2a-2) or (2a-3):



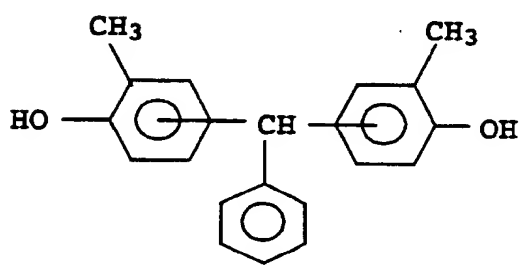
(2a-1)

wherein R is an alkyl group of 1-10 carbon atoms,



(2a-2)

wherein n is 1-6,



(2a-3)

The hardener represented by the molecular structure of the formula (2a-1) is a bifunctional hardener obtained by condensation reaction of phenol with a monoaldehyde obtained by substituting one hydrogen of formaldehyde with an alkyl group of 1-10 carbon atoms.

In the case of a combination of the bifunctional epoxy resin of the formula (1) with only the bifunctional phenolic hardener of the formula (2a-1), hardening is insufficient, and in order to obtain sufficient hardening, it is desirable to use a phenolic resin hardener other than the bifunctional hardener in combination with the bifunctional hardener of the formula (2a-1) which is used in an amount of 90% by weight or less.

R in the formula (2a-1) is an alkyl group of 1-10 carbon atoms, and if R is an alkyl group of 11 or more carbon atoms, fluidity at transfer molding decreases and moldability tends to deteriorate.

The hardener represented by the molecular structure of the formula (2a-2) is obtained by condensation reaction of phenol and benzaldehyde.

The hardener represented by the molecular structure of the formula (2a-3) is obtained by condensation reaction of o-cresol and benzaldehyde.

When other hardeners are used in combination with the hardener represented by the formula (2a) or (2b), such other hardeners include general polymers having mainly phenolic hydroxyl group. Examples of these hardeners are phenol novolak resins, cresol novolak resins, dicyclopentadiene-modified phenolic resins, copolymers of dicyclopentadiene-modified phenolic resins and phenol novolak resins or cresol

novolak resins, terpene-modified phenolic resins, p-xylene-modified phenolic resins and naphthol resins.

The inorganic fillers used in the present invention include, for example, fused silica powders, spherical silica powders, crystalline silica powders, secondary agglomerate silica powders, porous silica powders, silica powders obtained by grinding secondary agglomerate silica powders or porous silica powders and alumina. Especially preferred are fused silica powders, spherical silica powders and mixtures of fused silica powders and spherical silica powders. The amount of the inorganic filler is preferably 70-90% by weight based on the total amount of the composition from the point of balancing between soldering stress resistance and moldability.

The hardening accelerators used in the present invention may be any of those which accelerate reaction of epoxy group and hydroxyl group, and various accelerators which are generally used in sealing materials can be used. Examples thereof are diazabicycloundecene (DBU), triphenylphosphine (TPP), dimethylbenzylamine (BDMA) and 2-methylimidazole (2MZ). These may be used each alone or in combination of two or more.

The epoxy resin composition for sealing of the present invention comprises the epoxy resin, the hardener, the inorganic filler and the hardening accelerator as essential components. Furthermore, there may be optionally added various additives, for example, silane coupling agents, flame retardants such as brominated epoxy resins, antimony trioxide and hexabromobenzene, colorants such as carbon black and red iron oxide, releasing agents such as natural wax and synthetic wax, and low stress additives such as silicone oil and rubbers.

Molding materials can be prepared from the epoxy resin composition of the present invention by sufficiently uniformly mixing the epoxy resin, the hardener, the inorganic filler, the hardening accelerator and other additives by a mixer or the like, then further melt-kneading the mixture by hot rolls, kneader or the like, cooling the kneaded mixture and then grinding the mixture.

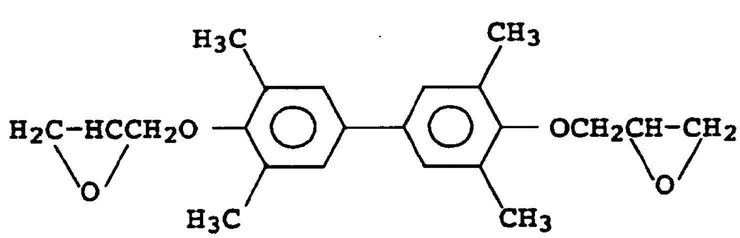
According to the present invention, epoxy resin compositions having soldering stress resistance which have not been able to be obtained by the conventional methods can be obtained and thus the resin compositions are considerably excellent in crack resistance when they undergo hot stress due to abrupt temperature change at soldering step and furthermore are superior in moisture resistance. Therefore, when the compositions are used for sealing, coating or insulating of electronic and electric parts, they are especially suitable for articles such as highly integrated large chip IC mounted on a surface mounting package which require very high reliability.

The following examples illustrate the present invention.

Example 1

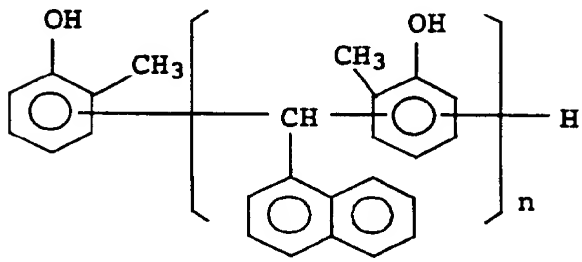
The following components were mixed at room temperature by a mixer, kneaded at 70-100 °C by a twin roll, cooled and ground to obtain a molding material.

Epoxy resin (softening point 110 °C, epoxy equivalent 190 g/eq) represented by the formula (1a)	3.8 parts by weight



(1a)

o-Cresol novolak epoxy resin (softening point 65°C, epoxy equivalent 200 g/eq)	8.7 parts by weight
Hardener (softening point 118°C, hydroxyl equivalent 180 g/eq) represented by the formula (2a-4)	2.3 parts by weight



(2a-4)

(a mixture comprising the compounds shown by n of 1-6 wherein the weight ratio is 1 of n=1, 0.2 of n=2, 0.1 of n=3 and 0.1 of n=4-6)

Phenol novolak resin hardener (softening point 90°C, hydroxyl equivalent 104 g/eq)	5.2 parts by weight
Fused silica powders	78.8 parts by weight
Triphenylphosphine	0.2 part by weight
Carbon black	0.5 part by weight
Carnauba wax	0.5 part by weight

The resulting molding material was shaped into tablets and chips of 6 × 6 mm were sealed in a 52p package by a low-pressure transfer molding machine under the conditions of 175°C, 70 kg/cm^2, 120 seconds for soldering crack resistance test, and chips of 3 × 6 mm were sealed in a 16p SOP package for soldering moisture resistance test. The sealed devices for testing were subjected to the following soldering crack resistance test and soldering moisture resistance test.

Soldering crack resistance test: The sealed devices for test were exposed to an atmosphere of 85°C, 85%RH for 48 hours and 72 hours and then dipped in a bath of solder of 260°C for 10 seconds. Thereafter, external cracks were observed by a microscope.

Soldering moisture resistance test: The sealed devices for test were exposed to an atmosphere of 85°C, 85%RH for 72 hours and then dipped in a bath of solder of 260°C for 10 seconds. Thereafter, they were subjected to pressure cooker test (125°C, 100%RH) and failure in opening of the circuit was measured.

The test results are shown in Table 1.

Examples 2-5

The components were mixed in accordance with the formulations of Table 1 and molding materials were prepared in the same manner as in Example 1. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 1.

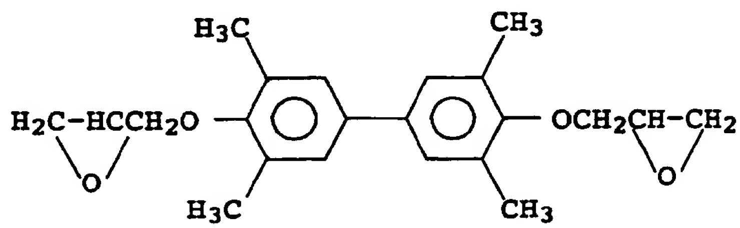
Comparative Examples 1-4

The components were mixed in accordance with the formulations of Table 1 and molding materials were prepared in the same manner as in Example 1. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 1.

Example 6

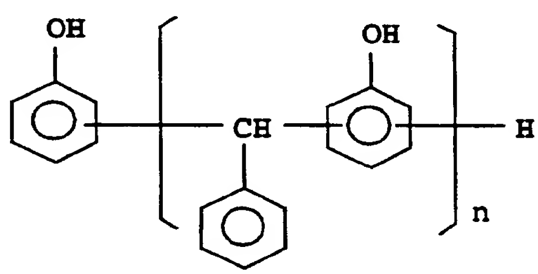
The following components were mixed at room temperature, kneaded at 70-100 °C by a twin roll, cooled and ground to obtain a molding material.

Epoxy resin (softening point 110 °C, epoxy equivalent 190 g/eq) represented by the formula (1a)	4.4 parts by weight



(1a)

o-Cresol novolak epoxy resin (softening point 65 °C, epoxy equivalent 200 g/eq)	8.1 parts by weight
Hardener (softening point 110 °C, hydroxyl equivalent 155 g/eq) represented by the formula (2a-2)	2.6 parts by weight



(2a-2)

(a mixture comprising the compounds shown by n of 1-4 wherein the weight ratio is 1 of n=1, 1.5 of n=2, 0.5 of n=3 and 0.1 of n=4)

Phenol novolak resin hardener (softening point 90 °C, hydroxyl equivalent 104 g/eq)	4.9 parts by weight
Fused silica powders	78.8 parts by weight
Triphenylphosphine	0.2 part by weight
Carbon black	0.5 part by weight
Carnauba wax	0.5 part by weight

Samples for testing were prepared using the resulting molding material and were tested in the same manner as in Example 1. The test results are shown in Table 2.

Examples 7-10

The components were mixed in accordance with the formulations of Table 2 and molding materials were prepared in the same manner as in Example 6. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 6. The results

are shown in Table 2.

Comparative Examples 5-7

The components were mixed in accordance with the formulations of Table 2 and molding materials were prepared in the same manner as in Example 6. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 6. The results are shown in Table 2.

Table 1

		Example					Comparative Example			
		1	2	3	4	5	1	2	3	4
Epoxy resin of the formula (1a)		3.8	10.2	11.3	7.3	7.0	2.6	12.4	3.1	0
o-Cresol novolak epoxy resin		8.7	0	0	3.1	4.6	7.9	0	9.4	13.2
Hardener of the formula (2a-4)		2.3	9.8	6.2	9.6	5.1	9.5	1.9	1.8	0
Phenol novolak resin hardener		5.2	0	2.5	0	3.3	0	5.7	5.7	6.8
Fused silica powder		78.8								
Triphenylphosphine		0.2								
Carbon black		0.5								
Carnauba wax		0.5								
Soldering crack resistance test (The number of samples having cracks/the total number of samples)	Moisture absorption 48 Hr	0/16	0/16	0/16	0/16	0/16	1/16	0/16	5/16	16/16
	Moisture absorption 72 Hr	0/16	0/16	0/16	0/16	1/16	7/16	4/16	14/16	16/16
Average life in soldering moisture resistance test (Hr)		>500	>500	>500	>500	>500	400	350	300	200

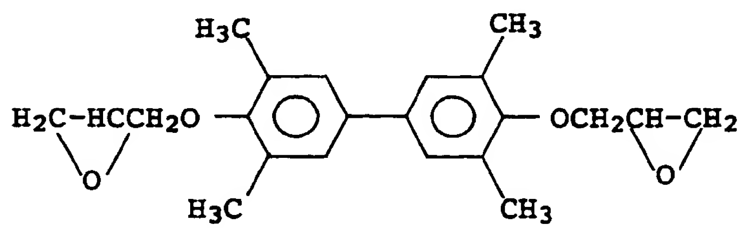
Table 2

		Example					Comparative Example		
		6	7	8	9	10	5	6	7
Epoxy resin of the formula (1a)		4.4	11.0	12.4	3.9	7.2	2.8	12.5	3.1
o-Cresol novolak epoxy resin		8.1	0	0	7.3	4.8	8.4	0	9.5
Hardener of the formula (2a-2)		2.6	9.0	2.7	8.8	4.8	8.8	1.8	1.8
Phenol novolak resin hardener		4.9	0	4.9	0	3.2	0	5.7	5.6
Fused silica powder		78.8							
Triphenylphosphine		0.2							
Carbon black		0.5							
Carnauba wax		0.5							
Soldering crack resistance test (The number of samples having cracks/the total number of samples)	Moisture absorption 48 Hr	0/16	0/16	0/16	0/16	0/16	2/16	3/16	1/16
	Moisture absorption 72 Hr	0/16	0/16	0/16	0/16	1/16	10/16	14/16	4/16
Average life in soldering moisture resistance test (Hr)		>500	>500	>500	>500	>500	340	300	360

Example 11

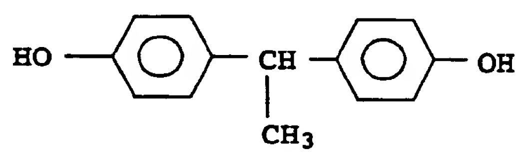
The following components were mixed at room temperature by a mixer, kneaded at 70-100 °C by a twin roll, cooled and ground to obtain a molding material.

Epoxy resin (softening point 110 °C, epoxy equivalent 190g/eq) represented by the formula (1a)	10.3 parts by weight



(1a)

o-Cresol novolak epoxy resin (softening point 65 °C, epoxy equivalent 200 g/eq)	2.6 parts by weight
Phenolic resin hardener (softening point 123 °C, hydroxyl equivalent 107 g/eq) represented by the formula (2a-1')	5.7 parts by weight



(2a-1')

Phenol novolak resin hardener (softening point 90 °C, hydroxyl equivalent 105 g/eq)	1.4 parts by weight
Fused silica powders	78.8 parts by weight
Triphenylphosphine	0.2 part by weight
Carbon black	0.5 part by weight
Carnauba wax	0.5 part by weight

The resulting molding material was shaped into tablets and chips of 6 × 6 mm were sealed in a 52p package by a low-pressure transfer molding machine under the conditions of 175 °C, 70 kg/cm^2, 120 seconds for soldering crack resistance test and chips of 3 × 6 mm were sealed in a 16p SOP package for soldering moisture resistance test.

The sealed devices for testing were subjected to the soldering crack resistance test and soldering moisture resistance test in the same manner as in Example 1.

The test results are shown in Table 3.

Examples 12-15

The components were mixed in accordance with the formulations of Table 3 and molding materials were prepared in the same manner as in Example 11. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 3.

Comparative Examples 8-11

The components were mixed in accordance with the formulations of Table 3 and molding materials were prepared in the same manner as in Example 11. Molded products for test were prepared by sealing

with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 3.

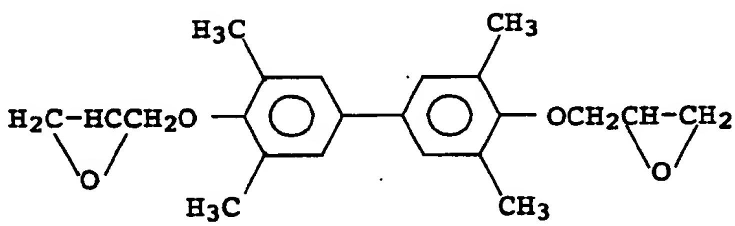
Table 3

		Example					Comparative Example			
		11	12	13	14	15	8	9	10	11
Epoxy resin of the formula (1a)		12.9	4.5	6.5	12.9	4.5	12.9	3.2	0	3.2
o-Cresol novolak epoxy resin		0	8.4	6.5	0	8.4	0	9.7	12.9	9.7
Phenolic resin hardener of the formula (2a-1')		6.0	6.0	3.5	2.5	2.5	1.8	6.0	7.1	1.8
Phenol novolak resin hardener		1.1	1.1	3.5	4.6	4.6	5.8	1.1	0	5.3
Fused silica powder		78.8								
Triphenylphosphine		0.2								
Carbon black		0.2								
Carnauba wax		0.5								
Soldering crack resistance test (The number of samples having cracks/the total number of samples)	Moisture absorption 48 Hr	0/16	0/16	0/16	0/16	0/16	1/16	3/16	8/16	16/16
	Moisture absorption 72 Hr	0/16	0/16	0/16	0/16	1/16	5/16	8/16	11/16	16/16
Average life in soldering moisture resistance test (Hr)		500<	500<	500<	500<	500<	280	350	300	100

Example 16

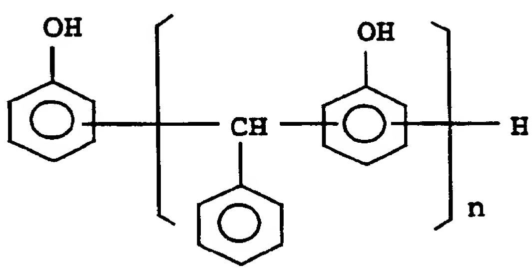
The following components were mixed at room temperature by a mixer, kneaded at 70-100 °C by a twin roll, cooled and ground to obtain a molding material.

Epoxy resin (softening point 110 °C, epoxy equivalent 190g/eq) represented by the formula (1a)	9.2 parts by weight



(1a)

o-Cresol novolak epoxy resin (softening point 65 °C, epoxy equivalent 200 g/eq)	2.3 parts by weight
Phenolic resin hardener (softening point 110 °C, hydroxyl equivalent 155 g/eq) represented by the formula (2a-2)	6.8 parts by weight



(2a-2)

(A mixture comprising the compounds shown by n of 1-4 wherein the weight ratio is 1 of n=1, 1.5 of n=2, 0.5 of n=3 and 0.1 of n=4)

Phenol novolak resin hardener (softening point 90 °C, hydroxyl equivalent 105 g/eq)	1.7 part by weight
Fused silica powders	78.8 parts by weight
Triphenylphosphine	0.2 part by weight
Carbon black	0.5 part by weight
Carnauba wax	0.5 part by weight

The resulting molding material was shaped into tablets, and chips of 6 × 6 mm were sealed in a 52p package by a low-pressure transfer molding machine under the conditions of 175 °C, 70 kg/cm^2, 120 seconds for soldering crack resistance test, and chips of 3 × 6 mm were sealed in a 16p SOP package for soldering moisture resistance test.

The sealed devices for testing were subjected to the soldering crack resistance test and soldering moisture resistance test in the same manner as in Example 1.

The test results are shown in Table 4.

Examples 17-20

The components were mixed in accordance with the formulations of Table 4 and molding materials were prepared in the same manner as in Example 16. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 4.

Comparative Examples 12-15

The components were mixed in accordance with the formulations of Table 4 and molding materials were prepared in the same manner as in Example 16. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 4.

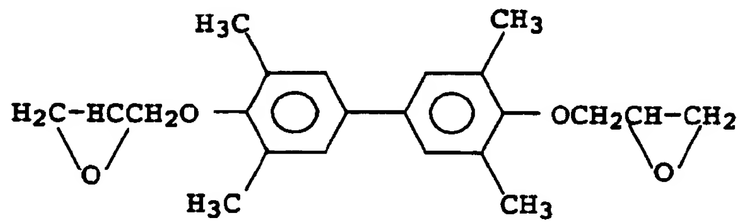
Table 4

		Example					Comparative Example			
		16	17	18	19	20	12	13	14	15
Epoxy resin of the formula (1a)		9.2	11.0	12.0	5.6	6.1	12.9	0	2.6	0
o-Cresol novolak epoxy resin		2.3	0	0	5.6	6.1	0	11.3	10.2	13.1
Phenolic resin hardener of the formula (2a-2)		6.8	9.0	4.0	8.8	3.9	0	8.7	1.4	0
Phenol novolak resin hardener		1.7	0	4.0	0	3.9	7.1	0	5.8	6.9
Fused silica powder		78.8								
Triphenylphosphine		0.2								
Carbon black		0.5								
Carnauba wax		0.5								
Soldering crack resistance test (The number of samples having cracks/the total number of samples)	Moisture absorption 48 Hr	0/16	0/16	0/16	0/16	0/16	2/16	2/16	6/16	16/16
	Moisture absorption 72 Hr	0/16	0/16	0/16	0/16	2/16	5/16	8/16	12/16	16/16
Average life in soldering moisture resistance test (Hr)		500<	500<	500<	500<	500<	300	420	280	100

Example 21

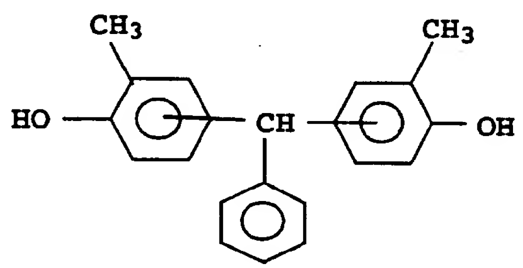
The following components were mixed at room temperature by a mixer, kneaded at 70-100 °C by a twin roll, cooled and ground to obtain a molding material.

Epoxy resin (softening point 110 °C, epoxy equivalent 190 g/eq) represented by the formula (1a)	9.3 parts by weight



(1a)

o-Cresol novolak epoxy resin (softening point 65 °C, epoxy equivalent 200 g/eq)	2.3 parts by weight
Phenolic resin hardener (softening point 90 °C, hydroxyl equivalent 152 g/eq) represented by the formula (2a-3)	6.7 parts by weight



(2a-3)

Phenol novolak resin hardener (softening point 90 °C, hydroxyl equivalent 105 g/eq)	1.7 part by weight
Fused silica powders	78.8 parts by weight
Triphenylphosphine	0.2 part by weight
Carbon black	0.5 part by weight
Carnauba wax	0.5 part by weight

The resulting molding material was shaped into tablets, and chips of 6 × 6 mm were sealed in a 52p package by a low-pressure transfer molding machine under the conditions of 175 °C, 70 kg/cm^2, 120 seconds for soldering crack resistance test, and chips of 3 × 6 mm were sealed in a 16p SOP package for soldering moisture resistance test.

The sealed devices for testing were subjected to the soldering crack resistance test and soldering moisture resistance test in the same manner as in Example 1.

The test results are shown in Table 5.

Examples 22-25

The components were mixed in accordance with the formulations of Table 5 and molding materials were prepared in the same manner as in Example 21. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 5.

Comparative Examples 16-19

The components were mixed in accordance with the formulations of Table 5 and molding materials were prepared in the same manner as in Example 21. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 5.

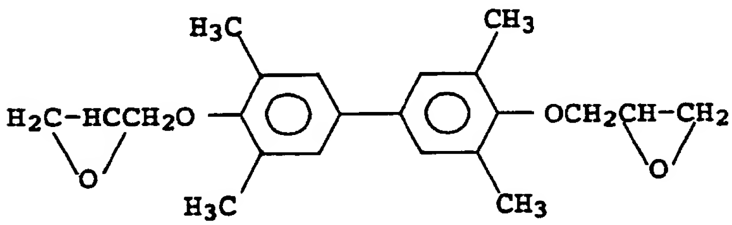
Table 5

		Example 21	Example 22	Example 23	Example 24	Example 25	Comparative Example 16	Comparative Example 17	Comparative Example 18	Comparative Example 19
Epoxy resin of the formula (1a)		9.3	8.9	12.0	5.6	6.1	12.9	0	2.6	0
o-Cresol novolak epoxy resin		2.3	2.2	0	5.6	6.1	0	11.4	10.2	13.1
Phenolic resin hardener of the formula (2a-2)		6.7	8.8	4.0	8.8	3.9	0	8.6	1.4	0
Phenol novolak resin hardener		1.7	0	4.0	0	3.9	7.1	0	5.8	6.9
Fused silica powder		78.8								
Triphenylphosphine		0.2								
Carbon black		0.5								
Carnauba wax		0.5								
Soldering crack resistance test (The number of samples having cracks/the total number of samples)	Moisture absorption 48 Hr	0/16	0/16	0/16	0/16	0/16	2/16	1/16	8/16	16/16
	Moisture absorption 72 Hr	0/16	0/16	0/16	0/16	0/16	7/16	8/16	14/16	16/16
Average life in soldering moisture resistance test (Hr)		500<	500<	500<	500<	500<	300	350	320	100

Example 26

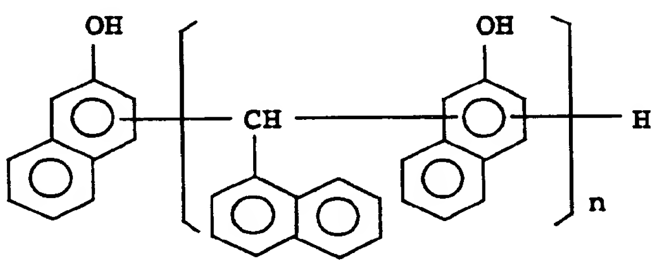
The following components were mixed at room temperature by a mixer, kneaded at 70-100 °C by a twin roll, cooled and ground to obtain a molding material.

Epoxy resin (softening point 110 °C, epoxy equivalent 190 g/eq) represented by the formula (1a)	6.8 parts by weight



(1a)

o-Cresol novolak epoxy resin (softening point 65 °C, epoxy equivalent 200 g/eq)	4.5 parts by weight
Hardener (softening point 114 °C, hydroxyl equivalent 216 g/eq) represented by the formula (2b-1)	5.3 parts by weight



(2b-1)

(a mixture comprising the compounds shown by n of 1-4 where the weight ratio is 1 of n=1, 0.2 of n=2 and 0.1 of n=3 or more)

Phenol novolak resin hardener (softening point 90 °C, hydroxyl equivalent 104 g/eq)	3.4 parts by weight
Fused silica powders	78.8 parts by weight
Triphenylphosphine	0.2 part by weight
Carbon black	0.5 part by weight
Carnauba wax	0.5 part by weight

The resulting molding material was shaped into tablets, and chips of 6 × 6 mm were sealed in a 52p package by a low-pressure transfer molding machine under the conditions of 175 °C, 70 kg/cm^2, 120 seconds for soldering crack resistance test and chips of 3 × 6 mm were sealed in a 16p SOP package for soldering moisture resistance test.

The sealed devices for testing were subjected to the soldering crack resistance test and soldering moisture resistance test in the same manner as in Example 1.

The test results are shown in Table 6.

Examples 27-30

The components were mixed in accordance with the formulations of Table 6 and molding materials were prepared in the same manner as in Example 26. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results

are shown in Table 6.

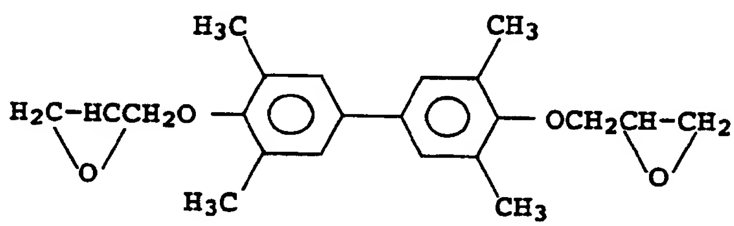
Comparative Examples 20-23

The components were mixed in accordance with the formulations of Table 6 and molding materials were prepared in the same manner as in Example 26. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 6.

Example 31

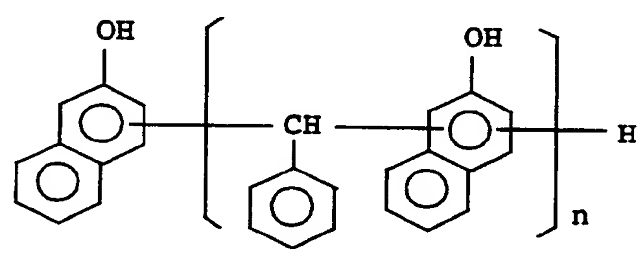
The following components were mixed at room temperature by a mixer, kneaded at 70-100 °C by a twin roll, cooled and ground to obtain a molding material.

Epoxy resin (softening point 110 °C, epoxy equivalent 190 g/eq) represented by the formula (1a)	3.8 parts by weight



(1a)

o-Cresol novolak epoxy resin (softening point 65 °C, epoxy equivalent 200 g/eq)	8.6 parts by weight
Hardener (softening point 109 °C, hydroxyl equivalent 200 g/eq) represented by the formula (2b-2)	2.3 parts by weight



(2b-2)

(a mixture comprising the compounds shown by n of 1-4 wherein the weight ratio is 1 of n = 1, 0.2 of n = 2, 0.1 of n = 3 or more)

Phenol novolak resin hardener (softening point 90 °C, hydroxyl equivalent 104 g/eq)	5.3 parts by weight
Fused silica powders	78.8 parts by weight
Triphenylphosphine	0.2 part by weight
Carbon black	0.5 part by weight
Carnauba wax	0.5 part by weight

Samples for the tests were prepared using the resulting molding materials in the same manner as in Example 26 and subjected to the tests in the same manner as in Example 1.
The test results are shown in Table 7.

Examples 32-35

The components were mixed in accordance with the formulations of Table 7 and molding materials were prepared in the same manner as in Example 31. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 7.

Comparative Examples 24-26

The components were mixed in accordance with the formulations of Table 7 and molding materials were prepared in the same manner as in Example 31. Molded products for test were prepared by sealing with the resulting molding materials. These molded products were subjected to the soldering crack resistance test and the soldering moisture resistance test in the same manner as in Example 1. The results are shown in Table 7.

Table 6

		Example					Comparative Example			
		26	27	28	29	30	20	21	22	23
Epoxy resin of the formula (1a)		6.8	9.4	12.1	2.9	3.7	2.3	12.3	3.1	0
o-Cresol novolak epoxy resin		4.5	0	0	6.6	8.6	7.2	0	9.3	13.2
Hardener of the formula (2b-1)		5.3	10.6	2.4	10.5	2.4	10.5	1.9	1.9	0
Phenol novolak resin hardener		3.4	0	5.5	0	5.3	0	5.8	5.7	6.8
Fused silica powder		78.8								
Triphenylphosphine		0.2								
Carbon black		0.5								
Carnauba wax		0.5								
Soldering crack resistance test (The number of samples having cracks/the total number of samples)	Moisture absorption 48 Hr	0/16	0/16	0/16	0/16	0/16	3/16	2/16	7/16	16/16
	Moisture absorption 72 Hr	0/16	0/16	0/16	0/16	1/16	7/16	7/16	12/16	16/16
Average life in soldering moisture resistance test (Hr)		>500	>500	>500	>500	>500	400	350	300	200

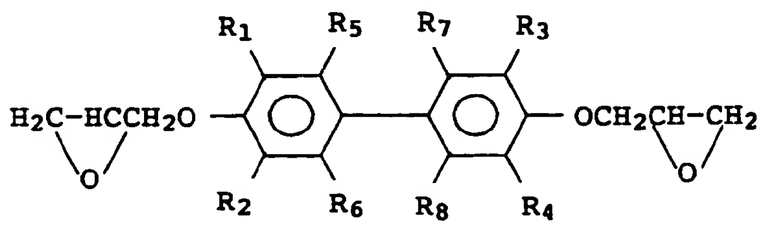
Table 7

		Example					Comparative Example		
		31	32	33	34	35	24	25	26
Epoxy resin of the formula (1a)		3.8	9.7	12.1	3.0	6.9	2.4	12.3	3.1
o-Cresol novolak epoxy resin		8.6	0	0	6.9	4.5	7.5	0	9.4
Hardener of the formula (2b-2)		2.3	10.3	2.4	10.1	5.2	10.1	1.9	1.8
Phenol novolak resin hardener		5.3	0	5.5	0	3.4	0	5.8	5.7
Fused silica powder		78.8							
Triphenylphosphine		0.2							
Carbon black		0.5							
Carnauba wax		0.5							
Soldering crack resistance test (The number of samples having cracks/the total number of samples)	Moisture absorption 48 Hr	0/16	0/16	0/16	0/16	0/16	5/16	4/16	7/16
	Moisture absorption 72 Hr	0/16	0/16	0/16	0/16	0/16	8/16	9/16	11/16
Average life in soldering moisture resistance test (Hr)		>500	>500	>500	>500	>500	340	300	360

Claims

1. An epoxy resin composition for sealing semiconductors which comprises as essential components:

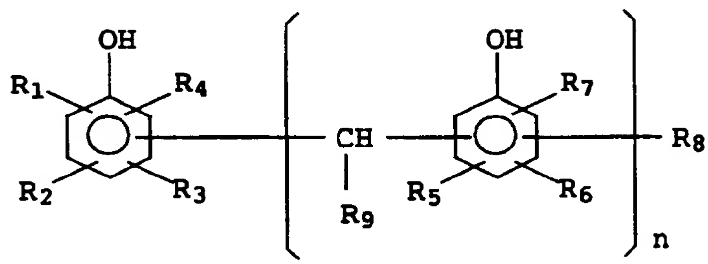
(A) an epoxy resin containing a biphenyl type epoxy resin represented by the following formula (1) in an amount of 30-100% by weight on the basis of the total amount of the epoxy resins,



(1)

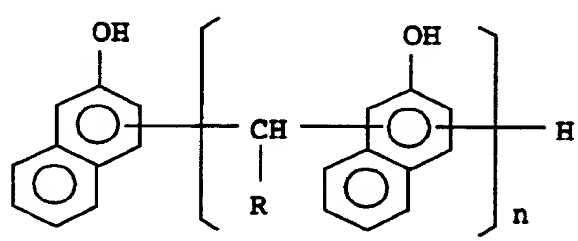
wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups,

(B) a hardener containing a phenolic resin hardener represented by the following formula (2a) or a naphthol resin hardener represented by the following formula (2b) in an amount of 30-100% by weight on the basis of the total amount of the hardener,



(2a)

wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups, R_9 is a group selected from phenyl, naphthyl, anthracenyl and alkyl groups, and n is 1-6,



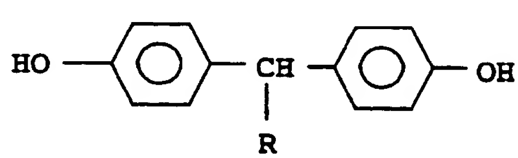
(2b)

wherein R is a group selected from phenyl, naphthyl, anthracenyl and alkyl groups and n is 1-6,

(C) an inorganic filler, and

(D) a hardening accelerator.

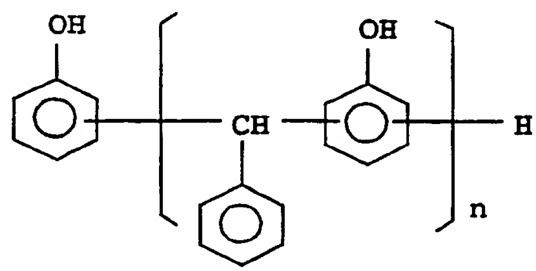
2. An epoxy resin composition according to claim 1, wherein the hardener (B) contains a phenolic resin hardener represented by the following formula (2a-1) in an amount of 30-90% by weight on the basis of the total amount of the hardener:



(2a-1)

wherein R is an alkyl group of 1-10 carbon atoms.

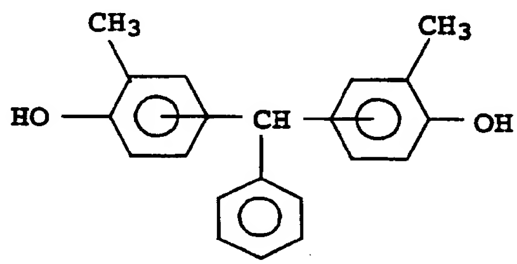
3. An epoxy resin composition according to claim 1, wherein the hardener (B) contains a phenolic resin hardener represented by the following formula (2a-2) in an amount of 30-100% by weight on the basis of the total amount of the hardener:



(2a-2)

wherein n is 1-6.

4. An epoxy resin composition according to claim 1, wherein the hardener (B) contains a phenolic resin hardener represented by the following formula (2a-3) in an amount of 30-100% by weight on the basis of the total amount of the hardener:



(2a-3)

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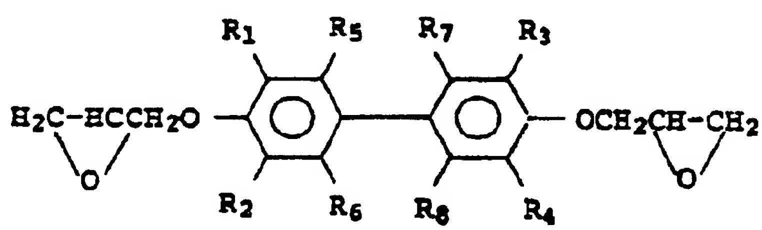
(54) **Epoxy resin composition based on diglycidylether of biphenyldiol.**

(57) Disclosed is an epoxy resin composition for sealing semiconductors which comprises as essential components:

(A) an epoxy resin containing a biphenyl type epoxy resin represented by the following formula (1) in an amount of 30-100% by weight on the basis of the total amount of the epoxy resin,



(1)

wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups,
(B) a hardener containing a phenolic resin hardener represented by the following formula (2a) or a naphthol

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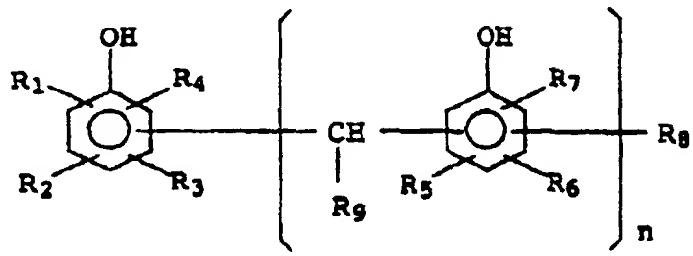
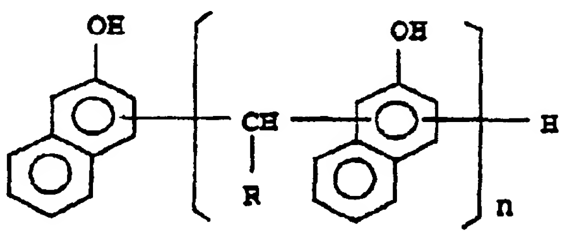
resin hardener represented by the following formula (2b) in an amount of 30-100% by weight on the basis of the total amount of the hardener,



(2a)

wherein R_1-R_8 are the same or different atoms or groups selected from hydrogen, halogens and alkyl groups, R_9 is a group selected from phenyl, naphthyl, anthracenyl and alkyl groups, and n is 1-6,



(2b)

wherein R is a group selected from phenyl, naphthyl, anthracenyl and alkyl groups, and n is 1-6,
(C) an inorganic filler, and
(D) a hardening accelerator.

European Patent Office

EUROPEAN SEARCH REPORT

Application Number
EP 93 10 7683

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.5)
P,Y	EP-A-0 536 748 (SUMITOMO CHEMICAL COMP., LTD.) * claims 1,4,7 * * page 3, line 29 - page 5, line 2 * * page 6, line 4 - page 8, line 2 * ---	1	C08G59/62 C08G59/24 H01L23/29 C08L63/02 C08L61/08 //(C08L63/02, 61:08), (C08L61/08, 63:02)
Y	DATABASE WPI Week 9216, Derwent Publications Ltd., London, GB; AN 92-126256 & JP-A-4 059 862 (TORAY IND. INC.) 26 February 1992 * abstract * ---	1	
P,A	EP-A-0 524 433 (DAI-ICHI KOGYO SEIYAKU CO., LTD.) * abstract; examples 4,11 * * page 3, line 21 - line 49 * * page 4, line 13 - line 49 * * page 6, line 2 - line 11 * ---	1	TECHNICAL FIELDS SEARCHED (Int.Cl.5)
A	PATENT ABSTRACTS OF JAPAN vol. 15, no. 484 (C-0892) 9 December 1991 & JP-A-03 210 322 (TOSHIBA CHEM. CORP.) 13 September 1991 * abstract * ---	1	C08G C08L H01L
A	DATABASE WPI Week 8528, Derwent Publications Ltd., London, GB; AN 85-168977 & JP-A-60 099 117 (SUMITOMO BAKELITE KK) 3 June 1985 * abstract * --- -/--	1-3	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
BERLIN	2 August 1994	Krische, D

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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EUROPEAN SEARCH REPORT

Application Number
EP 93 10 7683

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.5)
A	PROC.-ELECTRON. COMPON. TECHNOL. CONF., 42ND, 1992 pages 1023 - 1029 N. MOGI ET AL. * page 1023 - page 1024 * ---	1	
A	US-A-2 837 493 (F. SCHLENKER) * column 1, line 34 - column 3, line 3 * * example 3 * ---	1-4	
A	US-A-2 828 283 (M. DE GROOTE ET AL.) * claim 1 * * column 17, line 71 - column 23, line 34 * * column 35, line 74 - column 36, line 7 * * examples 13a,c13-21 * * examples c31-36 * ---	1-4	
A	US-A-2 499 370 (M. DE GROOTE ET AL.) * column 4, line 33 - column 5, line 39 * * column 14, line 62 - line 74 * -----	1	TECHNICAL FIELDS SEARCHED (Int.Cl.5)

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
BERLIN	2 August 1994	Krische, D

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